Fluorescence in damp air and comments on the radiative life time

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Photon yields in damp air excited by an electron using a ^{90}Sr β source are compared with those in dry air. Water vapors considerably reduce the yields, however, a further study is needed to evaluate the effects on the energy estimation of ultrahigh-energy cosmic rays. The relation of fluorescence efficiency to the life time of de-excitation by radiation is discussed.

1. Introduction

Photon yields in damp air are quite important to estimate the primary energy of cosmic rays with the fluorescence technique from space like EUSO. After our previous measurement in dry air [1, 2], the measurement has been continued using ^{90}Sr β source to study the pressure dependence of photon yields for radiation in damp air. The results in 21% and 56% relative humidities at one atmosphere are presented in section 3.

In our previous report [2], we were concerned with the decay time, τ. It consists of three terms: (1) the mean lifetime of the excited state to the lower state, τ_r, with which the fluorescence radiates; (2) that of internal quenching (internal conversion plus inter-system crossing) , τ_q ; and (3) the lifetime of collisional de-excitation, τ_c which is proportional to $\sqrt{T}/p$, where T and p are the temperature and the pressure, respectively. We have used the fluorescence efficiency; i.e. $\Phi(p) = \tau/\tau_r$, and $\Phi^\circ = \tau_o/\tau_r$, where τ_o is the combined lifetime of τ_r and τ_q .

Since Φ° was in order of 10^{-3} and τ_o was in order of a few tens ns, these led that τ_r would be in order of a few ten μs. This value of τ_r is quite large compared with ones obtained so far. Thus we will correct our relation between Φ° and the photon yield ϵ in the next section. We will discuss them in more detail in section **4**.

2. Photon yield

The photon yield per unit length per electron for the ith band at pressure p, $\epsilon_i(p)$ can be written by

$$\epsilon_i(p) = \rho\frac{\mathrm{d}E}{\mathrm{d}x}\left(\frac{1}{h\nu_i}\right) \cdot \varphi_i(p) \quad \text{, where } \varphi_i(p) = \kappa_i\,\Phi_i(p). \tag{1}$$

In eq.(1) $\Phi_i(p)$ is the fluorescence efficiency and we call $\varphi_i(p)$ the modified efficiency. The inverse of $\varphi_i(p)$ is expressed by

$$\frac{1}{\varphi_i(p)} = \frac{1}{\kappa_i\,\Phi_i^\circ}\left(1 + \frac{p}{p_i'}\right), \tag{2}$$

p_i' is the reference pressure where τ_{ci} is equal to τ_{oi} , namely $1/\tau_{ci} = (1/\tau_{oi})(p/p_i')$. $\Phi_i^\circ = \tau_{oi}/\tau_{ri}$ as before. κ_i in eq.(1) is introduced here different from refs [1, 2, 3]. Its physical meaning will be explained further in

section **5**. The eq.(1) can be written as a function of pressure as

$$\epsilon_i(p) = \frac{Cp}{1 + \frac{p}{p'_i}} \quad .$$ (3)

3. Experiment

The experimental details are described in [1, 2]. The central values of the narrow band filters used in the present measurement are 337.7, 356.3 and 392.0 nm and their bandwidths at half maximum are 9.8, 9.3 and 4.35 nm, respectively. The measurements have been done in dry air (mixing of 79% nitrogen and 21% oxygen) and in normal air with (21±4)% and (56±5)% relative humidities (RH) at one atmosphere. The pressure was varied from one atmosphere to 10 hPa under the temperature of (19.7±0.5)°C, keeping the specific humidity (SH : ratio of mass of the vapor to that of damp air) to be constant. The photon yield ϵ is determined from the number of signal counts, the total number of electrons, the length of the fluorescence portion, the solid angle of the photomultiplier(PMT), the quartz window transmission, the filter transmission and the quantum efficiency and the collection efficiency of the PMT.

The pressure dependence of ϵ in dry and damp air is shown in Figure 1 for three filter bands (We designate each filter band as 337, 358 and 391 nm). In these plots, the contribution of different bands in one filter band width are not separated, which is different from the results in our previous paper [1]. The results of least square(LS) fitting to the eq.(3) are shown by solid curves (dry), dashed curves (21%) and dotted curves (56%).
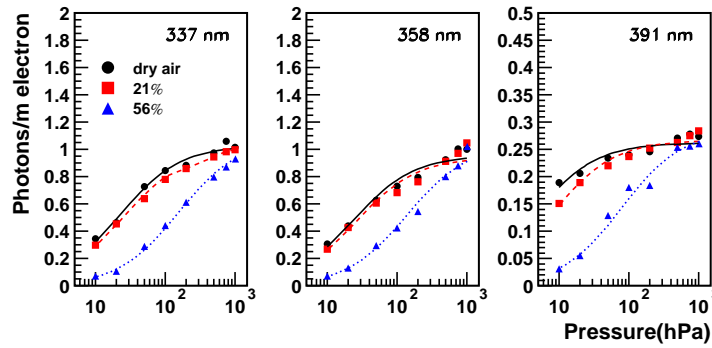

Figure 1. The pressure dependence of ϵ in dry air and damp air with RH, 21% and 56% at one atmosphere. The temperature is kept constant (19.7±0.5)°C throughout the measurements. In each series of measurements, SH (0, 3.0 and 8.0 g/kg, respectively) is kept constant, though RH changes.

The ratio of ϵ at SH=0, (3.0±0.5) and (8.0±0.7) g/kg to that of ϵ of dry air(SH=0) are plotted as a function of SH at 1000, 750 and 500 hPa in Figure 2. Calculated ratios with the LS fitted p' and C values determined above are shown by a solid, dashed and dotted curves for 1000, 750 and 500 hPa. The SH dependencies for 337 nm and 358 nm (2P transition) seem to be similar, but different from that for 391 nm (1N transition). We are repeating and continuing the measurements with different RHs to make the effect of water vapor more clear.
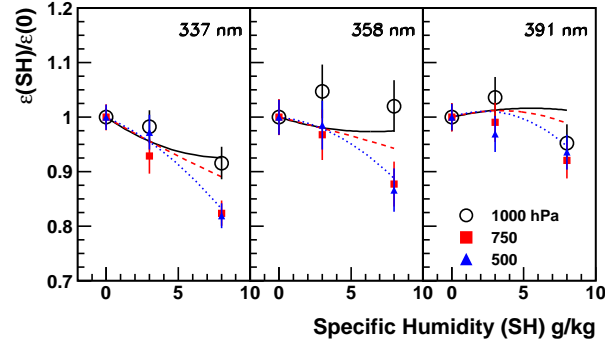
Figure 2. The ratio of ϵ at SH=0, 3.0 and 8.0 g/kg to that of ϵ of dry air(SH=0) are plotted as a function of SH at three pressures.

The reciprocal of the modified efficiency $\frac{1}{\varphi_i(p)}$ for air in $\frac{1}{\%}$ are plotted as a function of pressure in Figure 3. The results of LS fitting to eq.(2) are shown by solid (dry), dashed (21%) and dotted (56%) lines.
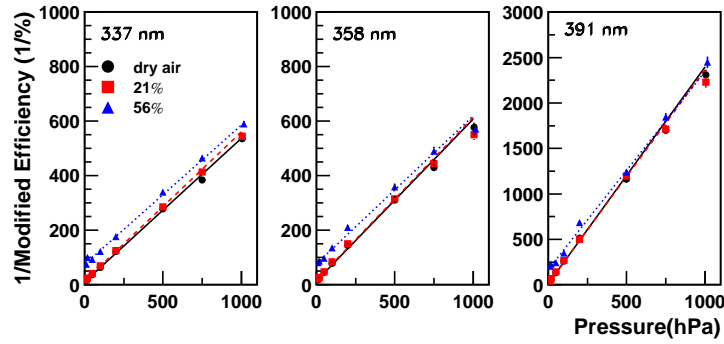

Figure 3. The reciprocal of the modified efficiency $\frac{1}{\varphi_i(p)}$ in dry and damp air are plotted as a function of pressure. The LS-fit lines are also shown.

In the present measurement the temperature was kept constant for decreasing the pressure. Actually, the atmospheric temperature decreases with the lapse-rate of about 6.5 K/km. The application of the present result to the real atmosphere is under study.

4.   Notes on radiative life time and photon yield of fluorescence

In eq.(1), a new parameter κ_i is introduced. If $\kappa_i = 1$, then $\varphi_i(p) = \Phi_i(p)$. In this case, eq.(1) holds only under very simple assumptions, namely (a) a charged particle is lost its energy dE by only one process, i.e. exciting an electron in molecule from the ground state to the only one excited state. And the excited electron emits $h\nu_i$ light or internally converts: $dE/h\nu_i$ represents the number of photons. Of course, this is not the case in air as well as N_2 and N_2^+.

The 2P transition does not take two steps, i.e. excitation and radiation. In 2P, the direct process from the ground state to the specific electronic state $C\,^3\Pi_u$ is forbidden because of the change of spin multiplet. There are many excited levels in N_2. And 2P radiative transition is not to the ground state. So the exciting energy is 11.03 eV while the radiation energy $h\nu_i$ is 3.68 eV for, say, 2P(0,0). While in 1N, nitrogen gas must be, of course, ionized at first. There are also many other excited electronic states of N_2^+ than $B\,^2\Sigma_u$. These facts lead that the simple assumptions mentioned above are not appropriate. The energy loss dE of charged particles may mainly be caused by ionization rather than excitation.

By referring Chapter 14 in [4], the factor κ_i in eq.(1) is inserted. This factor means the rate of the total energy loss dE with which an electron in a molecule is excited to the specific electronic state with a definite vibration state, such as $C\,^3\Pi_u$ electronic state with $v' = 1$ even indirectly. κ_i also contains a coefficient of proportion which may include the branching ratio of $B^{v',v''}$ [5], where v' and v'' are initial and final vibration levels, respectively, and the energy ratio of radiation to excitation and so on.

So, values of κ_i is considered to be much less than unity, say 10^{-3}. The values of Φ_i° in eq.(2) are in order of unity. Then the values of τ_r is the order of a few tens ns which is consistent with the old but more accurate τ_r for 2P [6, 7] and for 1N [7] than ours [2]. It is noted that the comments described here on τ_0 and τ_r do not have any effect on the analysis of the photon yield, ϵ.

5.   Conclusion

Photon yields in damp air under the constant temperature 19.7°C were investigated and compared with those in dry air. Though the rate of the mole number of water to the total mole number is small in damp air, water vapors seem to take a role in reducing the photon yields. We will continue to measure yields in damp air under various conditions.

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